**Institution: Newcastle University**

**Unit of Assessment: 10 Mathematical Sciences**

**Title of case study:** Keeping Track of Nuclear Fuel in Reprocessing

## 1. Summary of the impact

The Thermal Oxide Reprocessing Plant (THORP) at Sellafield in Cumbria has 20% of the world's current annual nuclear reprocessing capacity. Statistical methods developed in Newcastle during commissioning of THORP are integral to the nuclear material accountancy systems that are used in all stages of reprocessing. Since 2008 a Newcastle volumetric calibration system has been the only means of determining input into the plant. Regulatory approval of the system has ensured that THORP has been able to operate throughout the REF period, that customer costing has been accurate, and that the plant has complied with international standards for the close control of nuclear materials.

## 2. Underpinning research

The underpinning methodology for a volumetric system was developed by Henderson at Newcastle in 1993-1995, generalised in 1995-2000 in co-operation with McKnespiey (Newcastle) and Morton-Jones (Lancaster) and developed further with THORP operators British Nuclear Fuels Ltd (BNFL) from 2005. Recalibration was carried out in 2009/10 by Henderson in collaboration with Chater (BNFL, now Sellafield Sites) and Clarke (National Nuclear Laboratory). The methods have been on-going throughout the 2008-2013 period.

Operators and regulators use material accountancy methods to help safeguard against accidental or illicit diversion of nuclear materials. This requires highly accurate monitoring of all movements of nuclear material and regular inventories of storage. When, as at THORP, the materials are held and transferred in dissolved form, amounts of material can be estimated through a combination of sampling, chemical analysis, and either weight or volume determination, all of which are subject to error. A major statistical challenge during initial calibration is to determine the relationship between level and volume of liquor in a closed vessel containing significant interior pipework. The relationship is used to convert level measurements during operations to volume and in turn mass estimates of nuclear materials.

In generic statistical terms, volumetric calibration forms an inverse problem involving longitudinal data analysis with errors in variables. An unknown relationship $v=g(l)$ exists between volume and level in a vessel. The function $g(.)$ is continuous but at a finite but unknown number of points it is not differentiable. Smooth segments between discontinuities in first derivative may be linear or non-linear. During commissioning, p calibration runs are performed, in which a vessel is filled with measured aliquots of water from low to high volume, with $n_i$ aliquots in run i. Data ($V_{ij}$, $L_{ij}$) are available ($i=1,...,p$, $j=1,...,n_i$) subject to correlated error in $V_{ij}$ and heteroscedastic error in $L_{ij}$. The purpose is first to estimate g without bias for all l, and second to describe the associated error structure. Given a future measurement $L_0$, also subject to error, a prediction $V(L_0)$ is required together with a prediction interval.

The first challenge was design of calibration runs. Both volume and level are subject to measurement error. In reference [P1], theoretical expressions for mean square error were derived for competing methods. A technique based on in-tank density estimation was recommended on the grounds of robustness. This technique has been taken up by BNFL and continues to be used. In reference [P4] the stability of differential pressure measurements was studied, with a dynamic modelling approach recommended. A generic methodology paper [P6] shows that generalised least squares can be much less robust than ordinary least squares in the presence of measurement error, using calibration as an illustration.

The next decision was on aliquot sizes and number of runs. The first run begins with an empty vessel. Vessels cannot later be completely emptied: an unknown heel remains. Further, once active liquors are added to the vessel then access becomes impossible. Each calibration run is expensive (vessel capacity is up to 35 tonnes and a single run can take several days) meaning the number of runs needs to be limited. An expression for mean square error for the appropriate class of problem was derived in [P2].

The main methodology developed for fitting volume/level relationships was an early use of reversible jump Markov chain Monte Carlo [P3, P5]. A Bayesian approach was appropriate because prior (but unreliable) information from vessel design drawings is available. The underlying model for cross-sectional area is piecewise constant, leading to a piecewise linear relationship between level and volume, with prior information for slope changes. The Newcastle calibration methods were used during commissioning to calibrate over 100 process vessels within the THORP plant.

The calibration methods used bespoke Splus software written by Henderson and later developed into a commercial package based on the reversible jump methodology of Paul McKnespiey [P5], whose Newcastle PhD was partially funded by BNFL. The package was bought by Westlakes Research Institute for £45,000.

Recalibration of weighing systems is carried out periodically at THORP, using certified weights sequentially added to load systems. Recalibration of volume systems is more problematic; however, following necessary decommissioning of the weighing systems a bespoke validation analysis was undertaken in 2009/10 [E2, Section 5 below].

## 3. References to the research

*(ESARDA is the refereed proceedings of the biennial European Safeguards conference, the primary communication method for scientific developments in the area).*

[P1] Henderson, R., McKnespiey, P.N. and Temple, A. (1995). The volumetric calibration of tanks: design of trials. ESARDA, 17, 365-369.

[P2] McKnespiey, P.N. Henderson, R. and Temple, A. (1995). Volumetric calibration: use of in-tank density determination. ESARDA 17, 371-375.

[P3] Henderson, R., Temple, A. and McKnespiey, P. (1997). Computer intensive inference for calibration curves: experience at BNF THORP. ESARDA 19, 753-758.

[P4] Morton-Jones, A., Henderson, R., Hunt, B. and Binks, K. (1999). Optimal control of pressure measurements during volumetric calibration. ESARDA, 21, 285-289.

[P5] Henderson, R., Morton-Jones, A. and McKnespiey, P. (2000). Reversible jump MCMC for volumetric calibration. Journal of the Royal Statistical Society, Series C (Applied Statistics), 49 (4), pp. 563-576. ISSN 1467-9876 [* Key reference].

[P6] Morton-Jones, A. and Henderson, R. (2000). Generalized least squares with ignored errors in variables. Technometrics, 42(4), pp.366-375. DOI: 10.1080/00401706.2000.10485709. *(American Society for Quality and the American Statistical Association)*

## 4. Details of the impact

The amount of nuclear material transferred into and out of the THORP nuclear reprocessing plant in Cumbria is determined through the vessel calibration methodology developed in Newcastle and adopted by BNFL. THORP has the capacity to reprocess 1200 tonnes of nuclear fuel per year, which is 20% of the world's total current annual reprocessing capacity [E1]. Since 2008 the amount of nuclear material input into the plant has been determined exclusively through the level/volume methodology developed in Newcastle. Our methods have ensured that the plant can comply with regulator's requirements and has been able to operate throughout the REF period.

Originally a dual system (weight or level/volume) provided built-in redundancy for nuclear materials accountancy. In 2005 a pipe from a receipt vessel fractured in the containment area. Operations were suspended until 2008 and since restart only the level/volume systems have been available for determination of nuclear receipts. These rely entirely on the calibration methodology developed in Newcastle. The level/volume receipt systems were revalidated by Henderson and industry colleagues in 2009/10. Confidence in the systems was confirmed [E2] and operations at THORP have been allowed to continue.

1. Safety and environmental impacts

Close control of nuclear material is important to prevent the adverse safety and environmental impacts that would follow accidental release of radioactive material.

The 2005 leak was detected through nuclear material accountancy based on the vessel calibrations carried out using the Newcastle methodology. In [E3], Section 33, the Health and Safety Executive stated that "*The HSE investigation found that it was not the installed leak detection systems that led to the discovery of the leak. It was the analysis of nuclear materials accountancy (NMA) discrepancies at the end of several fuel shearing campaigns that led to the detailed investigations.... and subsequently to the discovery of the leak*."

2. Security impacts

Over 6000 tonnes of spent nuclear fuel from nine countries have passed through THORP calibrated vessels since commissioning. Close control of this fuel is extremely important for quickly identifying any security breaches or misuse of nuclear materials. The International Atomic Energy Agency (IAEA) is charged with monitoring civil programmes in order to ``*establish and administer safeguards designed to ensure that special fissionable and other materials ... are not used in such a way as to further any military purpose*" [E4].

One aspect of this safeguards work is material accountancy, by which records of movements of material are kept and regular inventories taken. The main receipt vessels at THORP are the Head End Accountancy Tanks (HEATs), which are used for input determination. The main issue vessels for plutonium product (HARPs) are used for output determination. Given their importance, the HEAT and HARP vessels were designed to have dual accountancy systems: a primary system based on weight and a secondary system based on volume/level. Highly accurate weighing systems were installed and calibrated using fairly straightforward models developed in Newcastle during 1993-1994. The more problematic and statistically challenging volumetric systems were installed, calibrated and recalibrated in 2009/10 using models described in the underpinning research above. Throughout, the European Atomic Energy Community (Euratom), which acts for IAEA in Europe, approved the methods and results [E5].

In [E6], the THORP materials custodian says "*The volumetric calibrations of the 12 main process vessels, which are also key measurement points for accountancy purposes, continue to be used* *on a daily basis and to pass the daily revalidation exercises*". The Newcastle work is key to this.

*3.* Commercial costing impacts

THORP is one of only three commercial light water reactor fuel reprocessing plants in the world [E7]. Customers are charged according to the quantities of material received, as measured in the receipt vessels, and THORP charges purchasing customers according to the amount of product, as measured at issue. The material of main interest is plutonium, which is generated during reprocessing of spent nuclear fuel. Extremely accurate measurement is necessary, with relative standard deviations of the order of 0.1% for plutonium being required. The Newcastle calibrations delivered this accuracy. Bias in the measurement system can lead to commercial charges measured in millions of pounds. Exact charges and costs are commercially confidential.

In summary, our volumetric system continues to be the sole means of determining transfers of nuclear materials into or out of THORP. Operations could have not have continued after 2008 without regulatory approval of these accurate and reliable accountancy systems.

**5. Sources to corroborate the impact**

[E1] "*Reprocessing Plants, Worldwide*", European Nuclear Society Encyclopaedia (2012). http://www.euronuclear.org/info/encyclopedia/r/reprocessing-plants-ww.htm, [accessed 04/09/2013]

[E2] Clarke, C.G, Chater, S.P. and Henderson, R. (2010). "*Review of HEAT A liquor measurement performance using volume density system"*. Sellafield Ltd Technical Report TTC/10/707 (N).

[E3] "*Report of the investigation into the leak of dissolver product liquor at the Thermal Oxide Reprocessing Plant (THORP), Sellafield, notified to HSE on 20 April 2005",* Health and Safety Executive (2007), available at: http://www.hse.gov.uk/nuclear/periodic-safety-review/thorp.htm. [accessed 04/09/2013]

[E4] "*IAEA Statutes Article III.A.5*", International Atomic Energy Agency (2013) http://www.iaea.org/About/statute.html, [accessed 04/09/2013]

[E5] Corroboration from the Head of Nuclear Fuel Cycle Analysis Section, Division of Information Management, Department of Safeguards, International Atomic Energy Agency & former Euratom inspector.

[E6] Corroboration from the Material Custodian, Thorp Chemical Plants, Sellafield Ltd, March 2011.

[E7] "*Processing of Used Nuclear Fuel*" (2012) World Nuclear Association, http://www.world-nuclear.org/info/inf69.html, [accessed 04/09/2013]